TEMPERATURE FIELDS IN APPARATUS FOR GROWING LEUCOSAPPHIRE SINGLE CRYSTALS BY USING MATHEMATICAL MODELING

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An approximate mathematical model and methodology for designing a horizontal directional crystallization (HDC) apparatus are proposed. The results of computations are compared with experimental data.

A method for the experimental investigation of the formation of fields T during the growing of leucosapphire single crystals is elucidated in [1], which permits obtaining reliable experimental data. Experimental results are presented there on the fields T directly in the technological process for one of the HDC apparatus. However, at this time there is already a large number of HDC units that differ in the structural solution and the geometric parameters of the crystallization and annealing zones. In this case it is not expedient to perform experimental investigations on each kind of apparatus. This is related primarily to the duration of the technological process (from 4 to 10-12 days). Moreover, existing equipment for conducting the experimental investigation should, as a rule, be subjected to a number of alterations, window installations in the screens and chamber, installation of additional measuring apparatus, etc., which cannot always be satisfied under factory conditions.

In this connection, the question of the development of a mathematical model which could be used to compute both the parameters of the technological process and the construction and parameters of the thermal component of the installation becomes no less urgent. In other words, there is a need to produce a mathematical model and a mode of computation for the class of installations as a whole. In this case experimental investigations are needed to establish the adequacy of the mathematical model to the real process in the installation, which in turn will permit the solution of two problems. On the one hand, this is the selection of the thermal regime required for growth by means of computing different versions on an electronic computer, and on the other, it is a sufficiently accurate computation of the structural elements of the thermal component, and a clarification of the degree of their influence on the melt–crystal system being formed in the technological process of the field T. This latter is also necessary for the development of equipment for another dimension of the crystal being grown, when the preliminary computations of the structural solutions reduce the design time significantly.

The present paper is devoted to the development of a method of designing an installation of the type SGVK-"Sapphire" intended for the growing of leucosapphire single crystals by the HDC method in order to obtain sufficiently confident quantitative results on the fields T in a leucosapphire at different stages of the technological process. The main attention in the development of the design and program method was turned to the possibility of designing the crystallization installation as a whole.

Such an approach naturally involves a whole set of simplifying assumptions in the computation of the field T in a leucosapphire; however, it affords the possibility of studying the influence of different structural elements of the installation (heaters, screens, diaphragms, etc.) on the field T being formed in the melt–crystal system by using the mathematical model. The relation between the field T and the energy parameters of the installation at different stages of the process also turns out to be quite important, since it permits the time program to control the heater power to be obtained in a first approximation.

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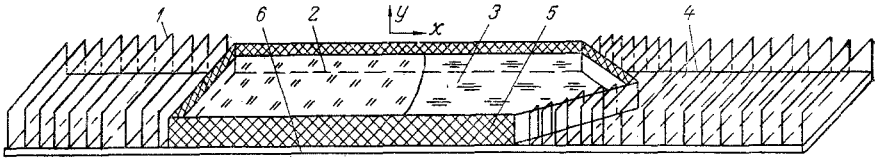

Fig. 1. Diagram of the working body of an installation for growing platelets of leucosapphire: 1, 4) bunches of molybdenum screens; 2) melt of initial material; 3) crystal to be grown; 5) container; 6) molybdenum stool.

However, the results obtained showed that the method and program developed permit quantitative estimation of the field T in the melt—crystal system also since comparing the computation and experiment results yields a discrepancy of not more than 3-4% in the absolute values of the temperature. Such accuracy is naturally small for a detailed study of the crystal growth process; however, it is totally adequate for the development of the thermal component of the apparatus in the design stage and permits dispensing with additional experiments on the model of the installation being developed for changes, say, in the size of the crystal to be grown.

In the formulation it is probably meaningful to separate the problem into "internal" describing the heat transfer directly in the container with the item, and "external" where the problem of heat transfer between the item, the heater, and the heat insulation, as well as in elements of the latter up to the cooled housing of the installation, is solved.

The container in which the crystallization is performed (Fig. 1) has the shape of a boat fabricated from molybdenum 5; thermally insulated at the endfaces by bunches of tungsten screens 1, 4; arranged on a common stool 6; at the center location with respect to the heater there is the crystallizing part of the material 3 and the melt 2.

Such a construction is a complex system, in the working space, which consists of several space domains, where each has its thermophysical and optical properties, dependent on the temperature and differing in direction.

Optical materials, such as leucosapphire, possess high transparency in a broad spectrum range, in which connection a radiation—conductive heat-transfer problem (RCHT) must be solved to find the field T. On the other hand, to analyze the growing process of single crystals, a two-dimensional problem must be solved as a minimum, however, the program to compute the RCHT has been worked out in a one-dimensional formulation for a plane material layer at this time [2, 3]. In this connection, an attempt is made in this paper to use a packet of different programs developed in *VNIIÉTO [4, 5] (in both the exact and approximate formulation) for the computation of the installation, which would permit development of a method of computation for similar installations and technological processes by reliance on experimental results [1, 6].

The computational diagram of the installation is presented in Fig. 2. The stool with the container in which the crystal growing process occurs is represented as a complex body consisting of several layers (I-IV) with different thermophysical and optical properties. The internal surface of the installation heating component is separated into 28 zones (numbers in circles) to take into account the most characteristic structure elements. The power is liberated in zones 5 and 24. The thermal insulation is a bunch of screens (the number of screens in the bunch is denoted by numbers in rectangles) arranged horizontally and vertically in different parts of the heating component, where the number of screens per bunch and the location correspond to the actual construction of the installation.

The temperature dependences of their thermophysical properties are given in solving the internal problem for each layer of complex load. The temperature field is described by a two-dimensional Fourier equation

$$c_p(T)\rho\frac{\partial T}{\partial \tau}=\frac{\partial}{\partial x}\left[\lambda_x(T)\frac{\partial T}{\partial x}\right]+\frac{\partial}{\partial y}\left[\lambda_y(T)\frac{\partial T}{\partial y}\right]. \quad (1)$$

The values of $C_p(T)\rho$ are given by different parabolic temperature dependences for the solid and liquid phases

*VNIIÉTO is the parent organization mentioned in the heading.

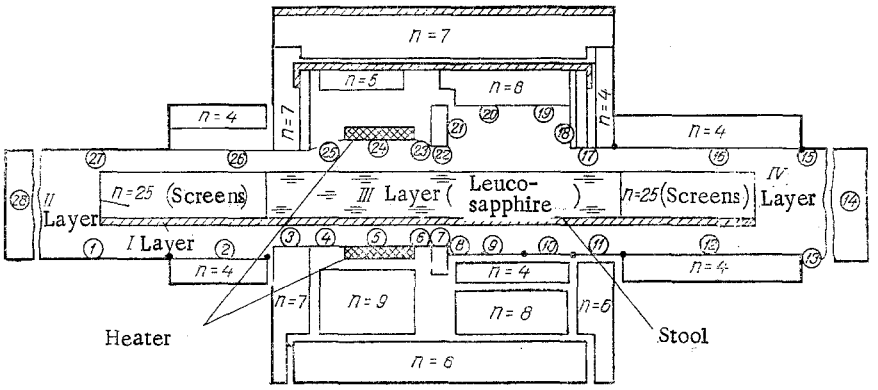

Fig. 2. Computational diagram of the installation (numbers in circles are the computation zones).

$$c_p\rho = \begin{cases} c_1\rho_1(T) & \text{at} \quad T < T_{pl}, \\ c_2\rho_2(T) & \text{at} \quad T \geqslant T_{pl}. \end{cases} \tag{2}$$

Such an approach permits analysis also of nonstationary processes, in particular, the influence of heater temperature fluctuations on the field T in the melt—crystal system and the position of the phase transition boundary. However, these questions are outside the scope of this paper and will be examined later.

The initial conditions are given either in the form of the initial temperature distribution over the item

$$T_{(x;\, y;\, \tau)} = T_{0(x;\, y;\, 0)}, \tag{3}$$

or the temperature at all points of the item is taken constant

$$T_{(x;\, y;\, \tau)} = T_0 = \text{const} \quad \text{at} \quad \tau = 0. \tag{3'}$$

In computing the nonstationary modes the Stefan conditions on the phase interface have the form

$$L\rho_p V = \lambda_s \left.\frac{\partial T_s}{\partial x}\right|_{x_p} - \lambda_l \left.\frac{\partial T_l}{\partial x}\right|_{x_p}, \tag{4}$$

where λ_s; λ_l are the heat conduction coefficients of the solid and liquid phases, respectively.

The condition of continuity of the heat flux (perfect contact) is satisfied on the boundaries between layers of the item with different thermophysical properties. Such an assumption can be considered valid since the initial material is melted and an almost perfect contact is formed with the bottom and walls of the container.

In the external problem the heat transfer by radiation between the item surface and the heat insulation and heater elements is computed by a zonal method. All the surfaces taking part in the heat transfer are separated into zones whose temperature is considered constant. The item is divided into 32 zones, of which 20 zones are on the leucosapphire. The conditions on the outer boundaries have the form

$$-\lambda_i \frac{\partial T}{\partial \bar{n}} = \frac{Q_i}{F_i}, \tag{5}$$

where λ_i is the heat conduction coefficint of the material in the i-th section.

The heat transfer by radiation is computed from the following equations

$$E_i\left(1 + \frac{\beta_i}{\mu_i}\right) - \sum_{j=1}^{m} \varphi_{j-i} E_j = f_i \quad (i = 1, 2, \ldots, m), \tag{6}$$

where E_i is the effective radiation flux from the surface of the i-th section onto the surrounding body, $\mu_i = \dfrac{1}{\varepsilon_i} - 1$; β_i is the coefficient for giving the resultant fluxes (β = 0)

or temperatures ($\beta = 1$) for sections on the item surface ($\beta = 1$); φ_{j-i} is the angular coefficient of heat transfer by radiation from the j-th to the i-th section, and $f_i = (1 - \beta_i) Q_i + 5.67 F_i \Theta_i$.

The heat fluxes through its elements that have sufficiently complex configuration (Fig. 2) are actually considered in computing the thermal insulation. The fundamental assumption taken is the absence of internal influence of the computation zones of thermal insulation on each other, i.e., the temperature field is considered one-dimensional and there is no heat flux by both heat conduction and radiation from zone to zone within them. The temperature dependence of their thermophysical properties are given for each of the layers and each heat insulation computation zone, where the number of layers and zones is not limited in principle and is determined by the possibilities of the electronic computer. This permits taking sufficiently accurate account of the real structural features of the apparatus. For instance, there can be in one heat insulation zone in the layers: screen insulation, backfill, felt, metal; in another, screens, refractory ceramic, metal; in the third, massive metal sheets, screens, backfill, etc. The one-dimensional Fourier equation

$$c_{m;\,n}(T)\,\rho_{m;\,n}\frac{\partial T}{\partial \tau} = \frac{\partial}{\partial x}\left[\lambda_{m;\,n}(T)\frac{\partial T}{\partial x}\right], \tag{7}$$

is used for the solution, where m;n is the number of the heat insulation computation zone and the layer, respectively, $C_{m;n}(T)$ is the temperature dependence of the specific heat in a given zone and layer, $\rho_{m;n}$ is the material density, $\lambda_{m;n}(T)$ is the heat conduction of the material of a given zone and layer. The number of equations equals the number of computation zones and layers.

In the case of screen heat insulation the number and material of the screens are modeled by using an equivalent heat conduction coefficient obtained from the flux balance for a system with a given quantity of screens N and a continuous heat insulation of identical thickness with a system of screens

$$\lambda_{eq} = \frac{\varepsilon_m \sigma_0 \delta}{N(2-\varepsilon_m)}(T_1 + T_N)(T_1^2 + T_N^2), \tag{8}$$

where ε_m is the emissivity of the screen material in the m-th zone, N is the number of screens in a given layer, and T_1 and T_N are the respective temperatures of the first and Nth screens.

Heat transfer from the cooling water or gas is accomplished on the cold heat insulation surface according to Newton's law

$$-\lambda_m \frac{\partial T_{m,\,n}}{\partial x} = (\alpha_{0;\,m} + \alpha_{1;\,m} T_{m;\,n})(T_{m;\,n} - T_{cold}); \tag{9}$$

where $\alpha_{0;m}$ is the coefficient of heat elimination to the water in the m-th zone, $\alpha_{1;m}$ is a constant governing the temperature dependence of $\alpha_{0;m}$, $T_{m;n}$ is the temperature in the m-th zone on the heat insulation boundary, and T_{cold} is the temperature of the cooling water or gas.

The initial conditions in the heat insulation have the form

$$T_{m;\,n}(x;\ \tau) = T_{m;\,n}(x;\ 0) \quad \text{or} \quad T_{m;\,n}(x;\ \tau) = T_0 = \text{const}. \tag{10}$$

In this connection, as already mentioned, the leucosapphire is a material partially transparent to thermal radiation: the exact solution of the two-dimensional internal problem (also complicated by the presence of a phase transition) on the basis of the RCHT equations becomes impossible. However, one of the most important questions for the constructor in designing and building the installation as well as in checking out the technological regime is the selection of the construction of the thermal component that will permit obtaining the field T in the item sufficiently close to that required by the conditions of the technological regime. As will be shown below, this problem can be solved successfully by using the considered approximate model of the installation, when complex RCHT computation programs are used as auxiliary in the material. In this case, concepts of effective heat conduction and equivalent emissivity of the material are used, which are computed by using programs for the exact solution of RCHT problems. Such an approach permits sufficiently good agreement between the computed results and experiment, and what is most important, the in-

vestigation of different versions of the structural solutions by using an electronic computer without relying on the production of a model of the installation to be developed.

The computation results obtained below afford the possibility of designing the structure of the thermal component that is sufficiently close to the optimal and corresponds to technology requirements in the preliminary stages of producing the installation. A seemingly fine tuning of the thermal component structure and of the technological regime is accomplished during the experiment, where different versions of the induced changes can also be computed first on the electronic computer to estimate the influence of these changes on the field T in the crystal.

The practical realization of the presented formulation of the problem is the following. To take account of internal heat transport in the leucosapphire because of RCHT, the possibility of giving the temperature dependence of the effective heat-conduction coefficient λ_{ef} as a fifth-degree polynomial in T is provided, where the polynomial coefficients are different in the x and y directions:

$$\lambda_x = \lambda_{0x} + \lambda_{1x}T + \lambda_{2x}T^2 + \lambda_{3x}T^3 + \lambda_{4x}T^4 + \lambda_{5x}T^5,$$
$$\lambda_y = \lambda_{0y} + \lambda_{1y}T + \lambda_{2y}T^2 + \lambda_{3y}T^3 + \lambda_{4y}T^4 + \lambda_{5y}T^5. \quad (11)$$

The majority of crystals of noncubic structure possess substantial anisotropy in the thermophysical properties, hence such accounting of the nonisotropic transport of the heat flux in the bulk of the growing material is practically always needed in a computation of the field T in crystals and opaque materials being grown with a definite orientation. In our opinion, this question is especially important when the results of solving the thermal problem are applied later to analyze thermoelastic stress fields. For semitransparent crystals, the values of λ_{ef} can differ in direction by more than one order when using the effective heat conduction approximation which depends on the thickness of the material layer and the boundary conditions in addition to the absorption coefficient.

Specific dependences for a leucosapphire crystal are computed as follows.

For a given container position relative to the heater, the length of the part being crystallized can be determined to 5-10% accuracy, hence the crystal thickness is known.

Furthermore, we assume that there is a plane layer of semiopaque material of thickness d, which is bounded by opaque walls with given temperatures T_1 and T_2 and a spectral emissivity $\varepsilon\lambda_1$ and $\varepsilon\lambda_2$. The temperature dependence of the thermophysical properties of the material is known. The temperature conditions (T_1) are given exactly on one of the boundaries (the crystallization front). This is the crystallization temperature, while the optical properties $\varepsilon\lambda_1$ are computed by starting from data on the refractive index and the absorption coefficient for the solid and liquid phases. On the other boundary, the spectral emissivity of molybdenum radiating into the leucosapphire is $\varepsilon\lambda_2$, while the temperature can be determined from a preliminary calculation in a first approximation, when λ_{ef} is computed by the Polets formula [7]. Moreover, we obtain the temperature distribution in a plane layer from the exact solution of the RCHT problem. The temperature dependence of λ_{ef} is then selected in such a way that the exact solution would agree with the approximate solution obtained with no worse than 0.5% error from the Fourier equation. The temperature dependence obtained for λ_{ef} is inserted into the main program, and a complete analysis of the installation is again performed, and the value of the temperature T_2 and the location of the crystallization front are refined. The selection of the dependence of λ_{ef} in a direction perpendicular to the direction of crystal growth is performed analogously, with the sole exception that a program with one contact and one open boundary is used for the exact RCHT solution. As practice has shown, the discrepancy in the temperature at the container nose after 3-4 iterations does not usually exceed 0.7%. For instance, for the middle position of the container relative to the heater, when the length of leucosapphire part being crystallized is 80 mm, the expression

$$\lambda_{ef\,x} = 4.446 \cdot 10^3 - 10.6T + 9.16 \cdot 10^{-3}T^2 - 3.37 \cdot 10^{-6}T^3 + 4.49 \cdot 10^{-10}T^4 \quad (12)$$

is obtained for the heat conduction along the x axis.

Temperature distributions in a 100-mm-thick leucosapphire layer are shown in Fig. 3 for the temperature drop T_1 = 2200°K, T_2 = 1000°K. It is well known that the known approximate methods [8, 9] which in the limit case of an optically thick layer, grazing radiation, yield

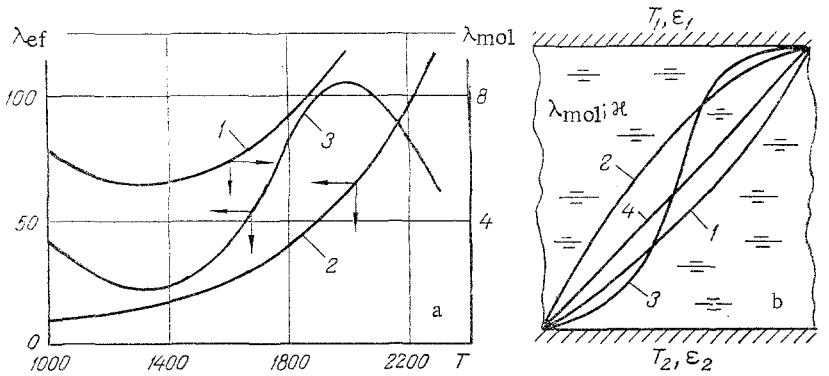

Fig. 3. Temperature dependence of the molecular and efficient heat conduction coefficients: a) computed dependences of λ_{ef} and the molecular heat conduction coefficient of leucosapphire from [9] [1) coefficient of molecular heat conduction, 2) computation of λ_{ef} by the Polets formula; 3) computation of λ_{ef} from the condition of agreement between the field T and the exact solution]; b) nature of the field T in a plane leucosapphire layer for different dependences of λ_{ef} on T [1) $\lambda_{ef} = \lambda_{mol}$, curve 1 in Fig. 3a; 2) λ_{ef} curve 2 in Fig. 3a; 3) λ_{ef} curve 3 in Fig. 3a, 4) λ_{ef} = const].

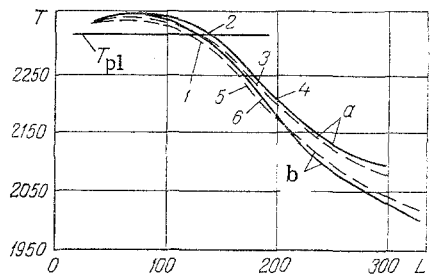

Fig. 4. Comparison of the computed results and the experimental data obtained by using an IR pyrometer: a) τ = 18 h, the field T at 18 h after the beginning of the process; b) τ = 23 h the same after 23 h. Values of the temperature gradients at the points mentioned: experimental: 1) 12-15 deg/cm; 3) 20-23; 5) 27-29; computed: 2) 14-15.5 deg/cm; 4) 22; 6) 32. T, °K; L, mm.

solutions which agree well with the exact value, are not applicable in this case (the limit conditions are not satisfied). The temperature dependence of λ_{ef} has a very complex form here (curve 3 in Fig. 3a), which however permits agreement between the results of the exact solution of the problem and computations from the Fourier equation. Since the method described is purely artificial, then the temperature dependence of λ_{ef} has no physical meaning.

Moreover, the effective emissivity ε_{ef} of the system (a leucosapphire layer of given thickness molybdenum substrate) is determined by the method of [10, 11] for the open leucosapphire surface in each of the computation zones, where the expression

$$\varepsilon_{\lambda_\perp} = (1 - R_\lambda)\left\{(1 - e^{-k_\lambda d})\left[1 + (1 - \varepsilon_\lambda^{n_{av}})e^{-k_\lambda d}\right] + \frac{\varepsilon_\lambda^{n_{av}}}{\pi} e^{-k_\lambda d}\right\}, \quad (13)$$

TABLE 1. Comparison of Results of Computation and Experiment

Container location in time (from beginning of the process), h	Temp. gradient on crystallization front, °K/cm			Temp. gradient in transition zone, °K/cm			Value of temp. in max. gradient domain, °K			Value of temp. at container nose, °K		
	computation	experiment	error, %	computation	experiment	error, %	computation	experiment	error, %	computation	experiment	error, %
τ=18	14	12	14,3	22	20	9,8	2226	2221	0,2	2090	2075	0,7
τ=23	15,5	15	3,3	32	29	9,35	2207	2197	0,4	1995	2017	1,1

is obtained for the normal spectral emissivity. Here $\varepsilon_\lambda^{n_{av}} = 1 - \frac{(n_m - n_{av})^2 + \varkappa_m^2}{(n_m + n_{av})^2 + \varkappa_m^2}$ is the spectral emissivity of the molybdenum substrate with radiation taken into account on the average with the refractive index n_{cp}; R_λ and k_λ are, respectively, the spectral coefficients of reflection and absorption, and $\varkappa_m$ is the index of absorption of the substrate material.

The integrated normal emissivity of the system is determined by integrating $\varepsilon\lambda_\perp$ with respect to the spectrum in the 0.2-30 mm range and is used in the computation of the radiant heat transfer in the working space of the installation.

The results of the computation show that [10, 11] ε_{ef} depends strongly on T, especially in the 1700-2000°K range, and on the thickness of the leucosapphire layer. Values of ε_{ef} for a system semiopaque material—metal substrate, are 1.7-2.2 times greater than for molybdenum, which indicates that utilization of the emissivity of molybdenum measured for radiation in a vacuum in the computations for an open surface can result in significant errors.

As an illustration of the complex analysis of a HDC installation, results on the field T in the direction of crystal growth are presented in Fig. 4 for two discrete container locations relative to the heater and are compared with the results of the experimental investigations presented in [6]. Good agreement is observed between the computed and experimental results (see table), while the qualitative pattern of the temperature distribution along the crystal length practically corresponds completely with experiment. This affords a foundation for considering that the influence of different structural elements of the installation (the quantity of screens and their disposition, the spatial location of the container and the heater, etc.) on the field T being formed can successfully be analyzed by using the method developed. Moreover, a computation of several discrete container locations with the crystal relative to the heater will permit determination the time program for a power change, in a first approximation, to realize the required temperature conditions which can be refined more finely during experimental checkout.

The complex experimental-design analysis performed for the apparatus to grow leucosapphire single crystals by the HDC method evidently showed that the main structural solutions of the working zone can be taken in the design stage on the basis of an analysis using the method proposed with a packet of computational programs. On the other hand, it is evident that a detailed quantitative analysis of the temperature conditions of semiopaque crystal growth can be performed only on the basis of the exact solution of the multidimensional RCHT problem or experimentally. Despite the utilization of a large number of different computational programs in this paper, the total computation time for one version is relatively small and is 25-40 min of machine time on a BÉSM-6 computer.

NOTATION

x, y, coordinates; ρ, material density; c_p, specific heat; τ, time; λx, λy, heat-conduction coefficients; λ, radiation wavelength; T, temperature; L, specific heat of a phase transition; ρ_p, density at the phase transition temperature; V, velocity of phase interface motion; x_p, phase interface coordinate; $\bar{n}$, normal direction; θ = T/100; Q_i, resultant flux; F_i, area of the radiant section; σ_0, Stefan—Boltzmann, constant; δ, thickness of the bunch of screens; α, heat elimination coefficient; ε, emissivity; n, refractive index; ϰ, index of absorption; k, absorption coefficient; d, thickness of the material layer.

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GROWTH OF A METAL ISLAND FILM UNDER CONDITIONS OF NON-STEADY-STATE REEVAPORATION OF METAL ADATOMS

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UDC 539.23

Taking into account the non-steady-state reevaporation of adatoms, the article solves the problem of diffusion growth of a system of uniformly distributed nuclei of the metallic phase. An expression describing the kinetics of change of the zone of nuclei capture is obtained.

Investigations of the kinetics of the growth of nuclei of the metallic phase showed that at the initial stages the changes in the nuclei are determined solely by the diffusion sink flow of adatoms [1]. Under certain conditions it is even possible that accumulations of atoms are displaced. These results enable us to view condensation at the stage of growth of an island film as a diffusion problem and to use the obtained analytical expressions for describing the kinetics of deposition of metallic films. Among the large number of works dealing with this problem we can distinguish two trends. One examines steady-state problems [2, 3]. The solution of the steady-state diffusion equation is applicable only in the calculation of systems for which the condensation time is much longer than the life of the adatoms in the adsorbed state. This state occurs at high temperatures of the surface of the substrate when the reevaporation of the metal adatoms is of steady-state nature. The works belonging to the second trend solve the non-steady-state diffusion equation [4, 5] but reevaporation of metal adatoms is not taken into account, i.e., the case is described when

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